

6.12 Electrochemical Advanced Oxidation

Natalia Sergienko, Wenli Jiang, Yanghua Duan, Andrew Haddad, b Ashok Gadgil,^{a,b} Jeffrey Urban,^b Baoxia Mi,^a and David Sedlak*^a

^a University of California, Berkeley, CA 94720

^b Lawrence Berkeley National Laboratory, Berkeley, CA 94720 Contact information: sedlak@berkeley.edu



Motivation

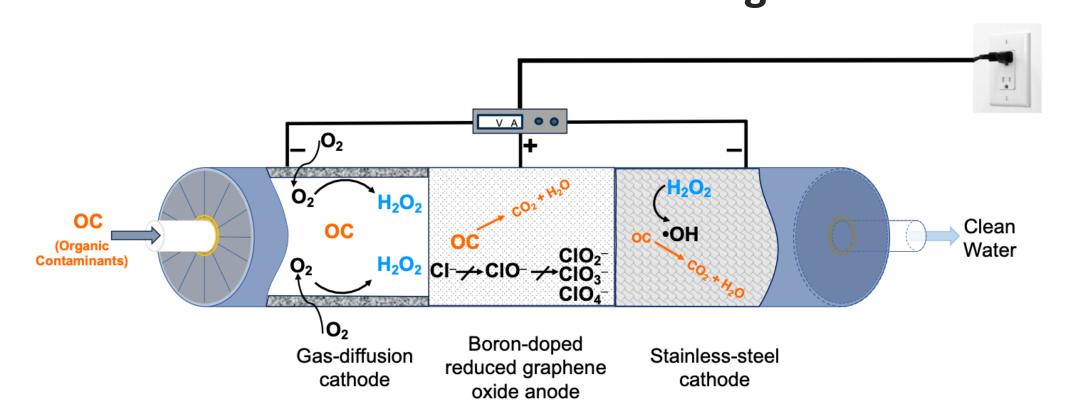
Treatment of trace organic contaminants is infeasible at small-scale due to challenges of replacing spent materials, an inability to remove certain contaminants, and economic feasibility.

Objectives

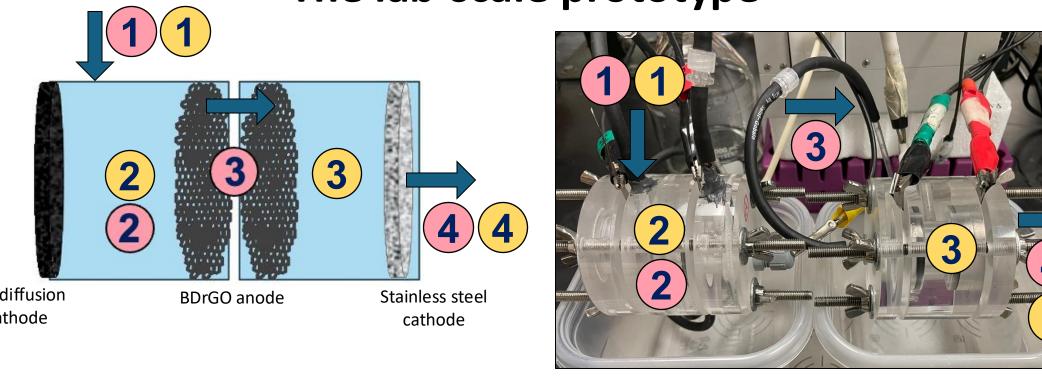
- Long-term operation of inexpensive electrodes with minimal maintenance
- Oxidation of contaminants with minimal production of toxic byproducts
- Autonomous operations that can adapt to variable influent water matrices

Approach

Electrified modular cartridge

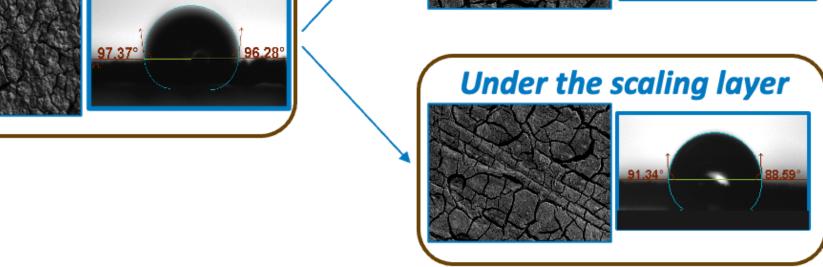


The lab-scale prototype

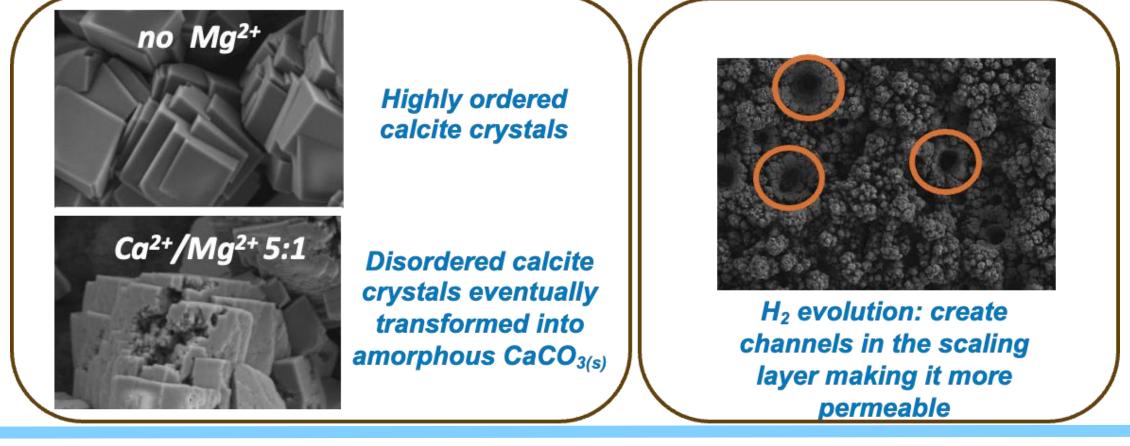


Gas-diffusion cathodes exhibit high stability

 Scaling prevents hydrophobicity loss In the absence of scaling Pristine catalyst



• Mg^{2+} and H_2 evolution create a mineral layer that does not lower efficiency $C/C_{Influent}$



pH regulation in the electrified cartridge

- The acidic pH entering the final cathode favored hydroxyl radical generation.
- No pH adjustment is needed, as the system naturally balances pH.

Experimental conditions: 50 mM Na₂SO₄ or SSW (20 μg/L contaminants) in a single-pass system (5 cm² electrodes, 0.5 mL/min) with 2–4 mA on the GDC and 1–3 mA on the stainless-steel frit cathode (-0.2 to -0.7 V vs Ag/AgCl).

Optimization of electrochemical control

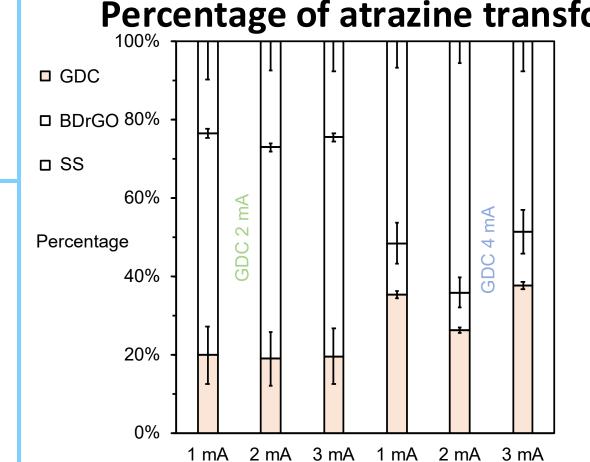
- Higher current boosts H₂O₂ production: 0.5 mM at 2 mA vs 1.6 mM at 4 mA.
- Faradaic efficiency improves: 40% at 2 mA \rightarrow 64% at 4 mA.
- Complete H₂O₂ utilization: No residual H₂O₂ in effluent.

Experimental conditions: same as above

• Higher current enhances removal efficiency: 4 mA on Gas Diffusion Cathode outperforms 2 mA.

- Atrazine removal improves: $^{55\%}$ at 2 mA \rightarrow $^{70\%}$ at 4 mA on Gas Diffusion Cathode
- Increased H₂O₂ production at 4 mA drives better contaminant removal.

Percentage of atrazine transformed



□Atenolol

▲ Ibuprofen

▲ Sulfamethoxazole

0.4 **-** □ Propranolol

SS cathode.

Current on SS cathode

Simulated surface water treatment

[CIO₃-] 2.0

•Optimized Conditions: 4 mA on Gas Diffusion Cathode for H₂O₂, 2 mA on

•Contaminant Removal: ~1-log in single-pass (~10s per electrode, SSW).

•Byproducts: No chlorinated byproducts above standards.

♦-2 mA+1 mA -♦-2 mA+2 mA

-**♦**-2 mA+3 mA -**▲**-4 mA+1 mA

-**△**-4 mA+2 mA -**△**-4 mA+3 mA

Concentration of H₂O₂

Transformation of Contaminants

□ Propranolol

■ Sulfamethoxazole

Atrazine

C/C_{Influent}

→2 mA+1 mA

-**♦**-2 mA+2 mA

-**◇**-2 mA+3 mA

△ 4 mA+1 mA

-**△**-4 mA+3 mA

□ Carbamazepin

■Ibuprofen

0 SSW

- SS cathode drives most removal (>50%) via ●OH from H₂O₂ activation.
- BDrGO anode is more effective at 2 mA on Gas Diffusion Cathode

EPA

30 [CIO₄-]

- 4 mA on Gas Diffusion Cathode improves overall removal despite lower efficiency of BDrGO anode.
- SS cathode: $1\rightarrow 2$ mA boosts removal by increasing •OH production, but 3 mA reduces efficiency as H₂O₂ is converted to H₂O instead of ●OH.

Capital and operating costs

- Capital cost: \$310, comparable to under-sink RO units (\$300-\$500).
- Low operating cost: \$0.0325/m³ for energy and \$67–100/year for maintenance.
- Scalable and efficient, offering a costeffective alternative to RO systems.

NAWI CONNECTIONS

Period of Performance: December 2024

Challenge Area/Topic Area: Materials & Manufacturing

Our project aims to develop inexpensive, robust, stable electrodes to enable pipe parity for non-traditional water sources through electrified, modular treatment.

NAWI Leverage

Our project leverages NAWI resources by integrating the KrCl reactor from Project 5.21* for UV treatment, building on prior advancements in Professor Gadgil's NAWI pilot

KEY FINDINGS AND CONCLUSIONS

Key Findings:

- Successful integration of three electrodes (GDC for H₂O₂ generation, BDrGO anode, SS cathode) into an electrified modular cartridge for efficient water treatment.
- Optimized design achieved >90% contaminant removal in a single pass, with high Faradaic efficiency, complete H₂O₂ utilization, and no need for pH adjustment.
- Scalable, cost-effective system (~\$309 capital cost), avoiding ion exchange membranes and minimizing byproduct formation, making it viable for distributed water treatment.

Conclusions: This project successfully developed a fully autonomous cost-competitive electrified treatment system with zero-chemical input for treatment of non-traditional water sources in small-scale applications.

REFERENCES

- Sergienko, N.; Bandaru, S.R.S.; Gadgil, A.J.; Sedlak, D.L. The Influence of Mg²⁺ on Scale Formation and Its Implications for Electrochemical H₂O₂ Generation by Gas Diffusion Cathodes. In preparation.
- Sergienko, N.; Haddad, A.; Kinigstein, E.; Sedlak, D.L. A New Perspective on the Role of Mg²⁺ in Calcite Electroscaling Formation. In preparation.
- Jiang, W.; Duan, Y.; Bandaru, S.R.S.; Radjenovic, J.; Sedlak, D.L.; Mi, B. (2023) Inhibition of Chlorinated Byproducts Formation by Boron-Doped rGO Electrodes During Electrooxidation of Trace Organic Contaminants. Applied Catalysis B: Environment and Energy, published online November 2024.
- Duan, Y.; Jiang, W.; Sedlak, D.L. (2023) Surface Processes Control the Fate of Reactive Oxidants Generated by Electrochemical Activation of Hydrogen Peroxide on Stainless-Steel Electrodes. Environmental Science & Technology, 58(47): 18680-18689.
- Duan, Y.; Sedlak, D.L. (2024) Electrochemical Hydrogen Peroxide Generation and Activation Using a Dual-Cathode Flow-Through Reactor: Enhanced Selectivity for Contaminant Removal by Electrostatic Repulsion. Environmental Science & Technology, 58(31): 14042–14051.

ACKNOWLEDGEMENTS

This material is based upon work supported by the National Alliance for Water Innovation (NAWI), funded by the U.S. Department of Energy, Energy Efficiency and Renewable Energy Office, Advanced Manufacturing Office under Funding Opportunity Announcement DE-FOA-0001905. Work at the Molecular Foundry was supported by the Office of Science, Office of Basic Energy Sciences, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.

